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An Evaluation of Potential Liner Materials for Eliminating FCCI in Irradiated Metallic Nuclear Fuel Elements

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Metallic nuclear fuels are being looked at as part of the Global Nuclear Energy Program for transmuting long-live transuranic actinide isotopes contained in spent nuclear fuel into shorter-lived fission products. In order to optimize the performance of these fuels, the concept of using liners to eliminate the fuel/cladding chemical interactions that can occur during irradiation of a fuel element has been investigated. The potential liner materials Zr and V have been tested using solid-solid diffusion couples, consisting of liner materials butted against fuel alloys and against cladding materials. The couples were annealed at the relatively high temperature of 700˚C. This temperature would be the absolute maximum temperature present at the fuel/cladding interface for a fuel element in-reactor. Analysis was performed using a scanning electron microscope equipped with energy-dispersive and wavelength-dispersive spectrometers (SEM/EDS/WDS) to evaluate any developed diffusion structures. At 700˚C, minimal interaction was observed between the metallic fuels and either Zr or V. Similarly, limited interaction was observed between the Zr and V and the cladding materials. The best performing liner material appeared to be the V, based on amounts of interaction.

I. INTRODUCTION

Metallic alloy fuels are being investigated for use in the United States Department of Energy (US-DOE) Global Nuclear Energy Program actinide transmutation program. Before being inserted into a reactor, the fuel alloys are jacketed with a stainless steel cladding. During irradiation, the fuel swells and contacts the stainless steel cladding. Once this happens, interdiffusion occurs between the fuel and cladding constituents, which is called Fuel Cladding Chemical Interaction (FCCI). As a result, the cladding constituents penetrate into the fuel and fuel constituents penetrate into the cladding. Based on available phase diagrams, relatively low melting point phases can form between U, Pu, and some fission products (e.g., the lanthanides) and the cladding constituents Fe and Ni.[1] Therefore, in terms of performance of the fuel in the reactor, it is of interest to totally eliminate the potential development of these low-melting phases due to FCCI.

One way to eliminate fuel-cladding interactions would be to employ a liner on the inner surface of a cladding tube. By using V or Zr, which have good neutronic characteristics, no low melting phases would form between the fuel or fission products and the cladding constituents, based on binary phase diagrams. In order to study how a metallic fuel alloy and different cladding alloys would interact with V and Zr, diffusion couple experiments have been performed at 700˚C using a cast U-based alloy with added fission products (to imitate high-burn-up fuel) and three cladding alloys. The fuel alloy contained U, Pu, and Zr as the main constituents and minor additions of Nd, Mo, and Ru. This paper will describe the results from these experiments. Comments will be made about the rate of interaction between the V and Zr and the metallic fuel and between the V and Zr and the cladding materials. Additionally, the morphology of the developed diffusion structures will be described. Comments will also be made about the potential of using V or Zr as a barrier material.

II. EXPERIMENTAL

One fuel composition was employed for the diffusion couple experiments, viz. 65U-19Pu-9Zr-2Nd-2.5Mo-2.5Ru[a]. This alloy imitates a high burn-up fuel. The Nd addition represents the relatively large concentrations of lanthanide fission products present in such a fuel, and the Mo and Ru represent the noble metal fission products that would be present. Lanthanide and noble metal components have been shown to participate in fuel-cladding interaction to form phases in actual irradiated fuels.[2] For the steels, a HCM12A alloy[b] and two oxide dispersion strengthened (ODS) steels[c] were used. These claddings represent "advanced" cladding alloys that are being investigated due to their potentially improved irradiation performance. Diffusion couple "stacks" were assembled by alternating liner and fuel and liner and cladding samples. These "stacks" were pressed and clamped together in a Kovar steel jig, which consisted of two end plates and three threaded Kovar rods. Kovar steel has a low coefficient of thermal expansion. The diffusion couple assemblies were wrapped together with some Zr pieces (acting as an oxygen getter) in Ta foil and heated to

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[a] All compositions are in weight percent.

[b] The HCM12A alloy had a composition of: 10.54Cr-0.98Cu-0.63Al-0.62Mn-0.54Nb-0.39Ni-0.32Mo-0.32O-0.28Si-0.19V-0.15Si-0.11C-balFe.

[c] One ODS steel (Alloy 1) was a research alloy and had a composition of: 10.78Cr-7.72Mo-0.46Ti-0.10Y_2O_3-0.0622O-0.053C-0.02Si-0.015Mn-0.009N-0.001S-balFe. The other was alloy MA957, which had a composition of: 10.83Cr-1.02Cu-0.64Mn-0.30Mo-0.27Si-0.19V-0.11C-0.0063N-0.0054Nb-0.0016P-0.0002S-balFe.

temperature in a manually controlled furnace located within an Ar atmosphere glovebox. Samples were annealed for 50, 75, or 200 hours at a temperature of 700˚C, which is considered the absolute maximum temperature the fuel/cladding interface could be exposed to at the most aggressive operating conditions. After annealing, the samples were rapidly cooled within 20 minutes using flowing He. The annealed samples were encased in epoxy, sliced longitudinally using a slow-speed saw to expose the fuel-steel interface and direction of diffusion and then polished (grinding papers through 1200 grit). Microstructural analysis was performed on the diffusion couples using a Zeiss 960A scanning electron microscope (SEM). Elemental compositions and spatial distributions were obtained with an Oxford Energy Dispersive Spectroscopy (EDS) and Wavelength Dispersive Spectroscopy (WDS) spectrometer equipped with ISIS LINK software.

III. RESULTS

III.A. Liner Versus Fuel Couples

Two diffusion couples were annealed at 700˚C for 75 hours between the fuel alloy and the liner materials: one was mated against V, the other against Zr. Figure 1 shows the microstructure of the unreacted fuel after this annealing treatment. The fuel alloy is multiphase. The fission product Nd is contained in a separate precipitate phase vis-à-vis the Mo and Ru. The diffusion structures that developed for the two diffusion couples are presented in Figure 2. Based on the morphology of the interdiffusion zones, it can be seen that the overall interdiffusion zone width is larger for the couple with Zr than for the one with V. The interdiffusion zone for the couple with V seems multi-phase, and the couple with Zr seems single phase. No evidence of cracking or sample fallout during polishing is apparent. So, it appears that brittle phases do not form in the interdiffusion zones.

As shown in Table 1, the couple with Zr exhibits an interdiffusion zone around 120 μm wide, while the V couple has an interdiffusion zone around 10 μm wide. X-ray maps were generated for each diffusion couple to determine how the various fuel and barrier material constituents were distributed in the interdiffusion zones (see Figs. 3 and 4). Based on these X-ray maps, it can be seen that Nd is the only constituent not observed at a noticeable concentration in the interdiffusion zone of the fuel versus Zr couple. For the fuel versus V couple, higher magnification X-ray maps were produced to confirm the observation that negligible U, Pu, and Ru were observed in the interdiffusion zone. In this couple, Pu-rich particles were observed in the V, and they seem to be on the surface of the sample and may have been a result of sample polishing.

III.B. Liner Versus Cladding Alloy Couples

The interaction zones that form between V and Zr and ODS alloy 1, MA957, and HCM12A after different annealing treatments at 700˚C are presented in Figs. 5-7. For ODS alloy 1, MA957, and T122, there is less reaction with V than there is with Zr. The various interdiffusion zone widths for the annealed diffusion couples are enumerated in Table 1.

With respect to the relative compatibility of the various cladding alloys with V, MA957 exhibits a 10-μm-wide interdiffusion zone after 200 hours at 700˚C, while for ODS alloy 1 the interdiffusion zone is at 10 μm after only 75 hours. Therefore MA957 exhibits better compatibility with V. The compatibility of alloy HCM12A with V seems analogous to what is observed for MA957.

With respect to compatibility with Zr, the couples with alloy HCM12A and MA957 exhibit interdiffusion zones of similar width after 200 hours at 700˚C. After 50 hours at 700˚C, alloy HCM12A reacts less with Zr than does ODS alloy 1. Therefore, overall HCM12A and MA957 are the most compatible alloys with Zr.

IV. DISCUSSION

Based on the results discussed above, both V and Zr appear to be good candidate materials to serve as barriers to FCCI in irradiated metallic fuels. The interdiffusion zones that form for the fuel/V and fuel/Zr couples and for V/cladding and Zr/cladding couples are relatively small compared to what has been reported for couples run at 700˚C between a 65U-19Pu-9Zr-2Nd-2.5Mo-2.5Ru alloy and various cladding alloys.[3] In couples between the fuel alloy and ODS and HCM12A claddings annealed at 700˚C for 25 hours, the widths of the interdiffusion zones were 400 and 525 μm, respectively. In the fuel/cladding couples, not only were interdiffusion zones wide but there was evidence that some phases in the formed interdiffusion zones had melted, and some phases were brittle and had cracked during sample polishing. No signs of melting were observed in any of the diffusion couples reported in this study, and there were no signs of cracking or sample pullout during polishing. Mechanical integrity is a very important attribute of a liner material. A liner must remain robust during service. This means that not only should there be limited interaction with the cladding and fuel, but any interaction that does occur should not result in the formation of brittle phases. The interdiffusion zones that formed in the couples reported in this investigation are likely to exhibit acceptable mechanical properties.

V. CONCLUSIONS

Diffusion couples were annealed for various times at 700˚C between the two potential barrier materials V and Zr and a 65U-19Pu-9Zr-2Nd-2.5Mo-2.5Ru alloy and three cladding alloys (ODS alloy 1, HCM12A, and MA957), and based on the results of these couples the following conclusions can be drawn:

1. Even at the relatively high temperature of 700˚C, V and Zr exhibit attributes that make them good candidate barrier materials for use in metallic fuel elements: good chemical compatibility with prototypic cladding materials and good compatibility with a metallic fuel alloy that imitates high burnup fuel.

2. V interacts less with cladding and fuel alloys in diffusion couple tests run at 700˚C than does Zr.

3. Brittle or low-melting phases do not form in interaction zones between fuel or cladding alloys and V or Zr.

ACKNOWLEDGMENTS

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Table I. Size of Interaction Zones for Diffusion Couples Annealed at 700˚C.

Couple	Barrier Material	Cladding Alloy	Fuel Alloy	Time (Hrs)	Interaction Zone Width (μm)
1	V	HCM12A	-	50	3
2	V	ODS	-	50	2
3	V	ODS	-	75	10
4	V	MA957	-	200	10
5	V	HCM12A	-	200	10
6	Zr	ODS	-	50	5
7	Zr	HCM12A	-	50	Negligible
8	Zr	MA957	-	200	15
9	Zr	HCM12A	-	200	15
10	V	-	65U-19Pu-9Zr-2Nd-2.5Mo-2.5Ru	75	10
11	Zr	-	65U-19Pu-9Zr-2Nd-2.5Mo-2.5Ru	75	120

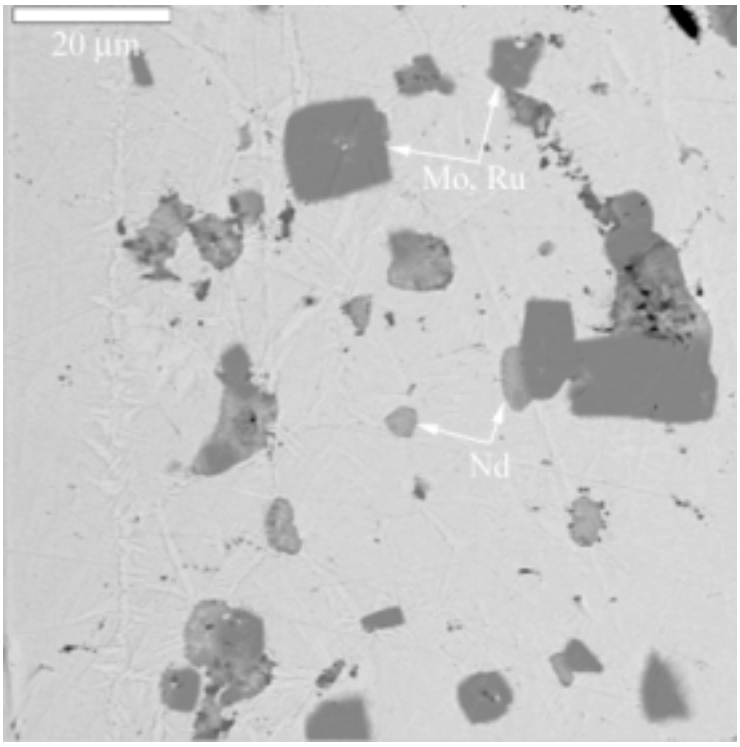
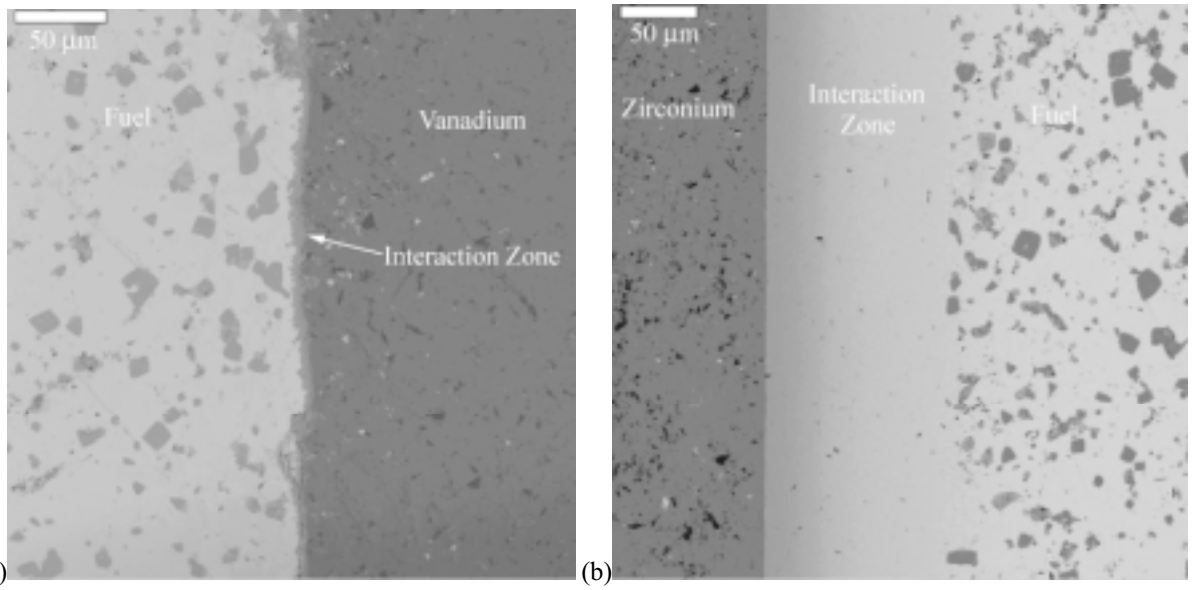


Fig. 1. SEM micrograph of the as-cast 65U-19Pu-9Zr-2Nd-2.5Mo-2.5Ru alloy that had been annealed at 700˚C for 75 hours.



(a) (b)

Fig. 2. SEM micrographs showing the diffusion structures that developed for the couples between a fuel doped with fission products and (a) V and (b) Zr when they were annealed at 700˚C for 75 hours. The interaction zone forms between the fuel and the V or Zr.

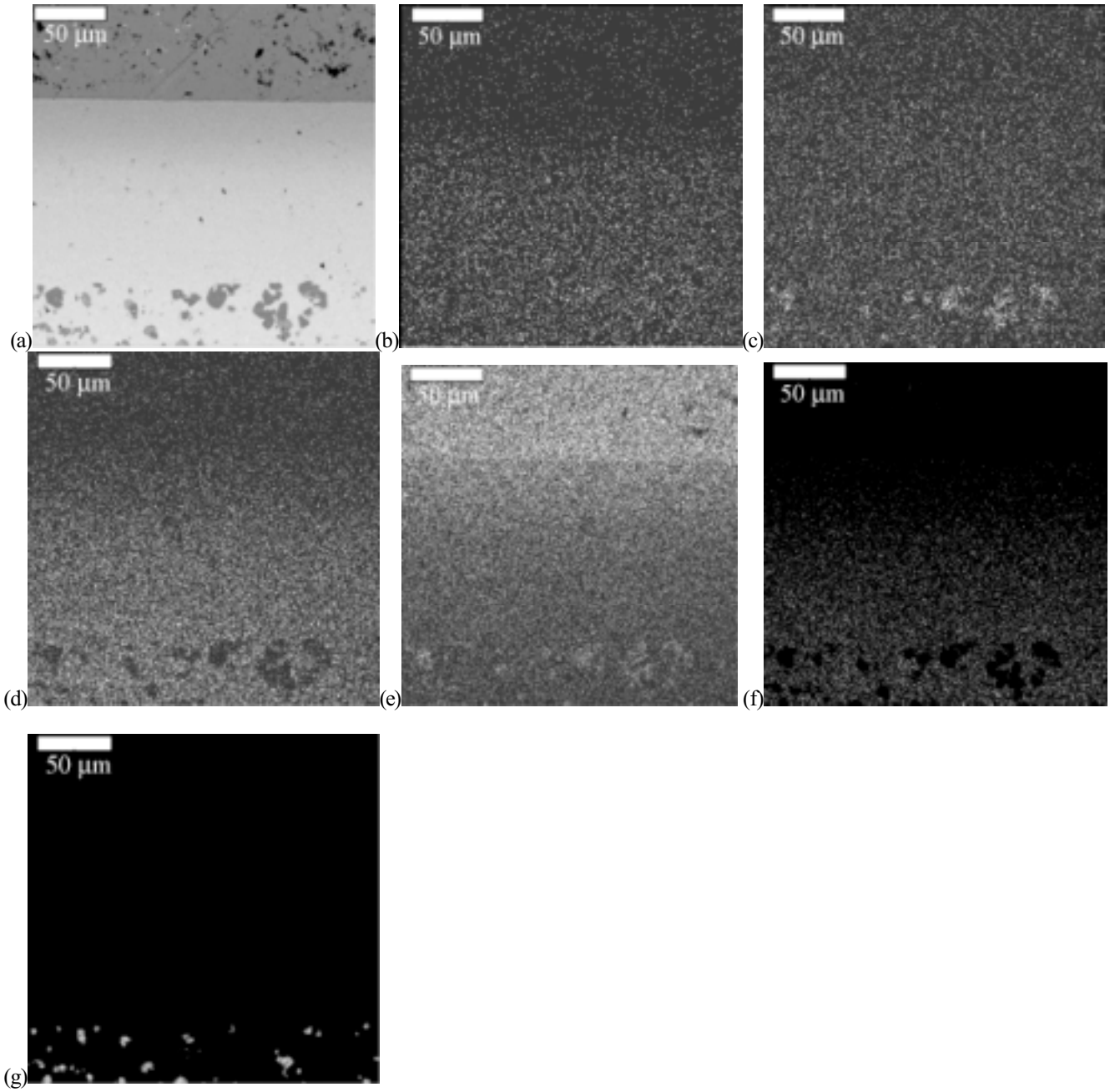


Fig. 3. Backscattered electron image (a) and X-ray maps for (b) Mo, (c) Ru, (d) U, (e) Zr, (f) Pu, and (g) Nd for the Fuel versus Zr couple.

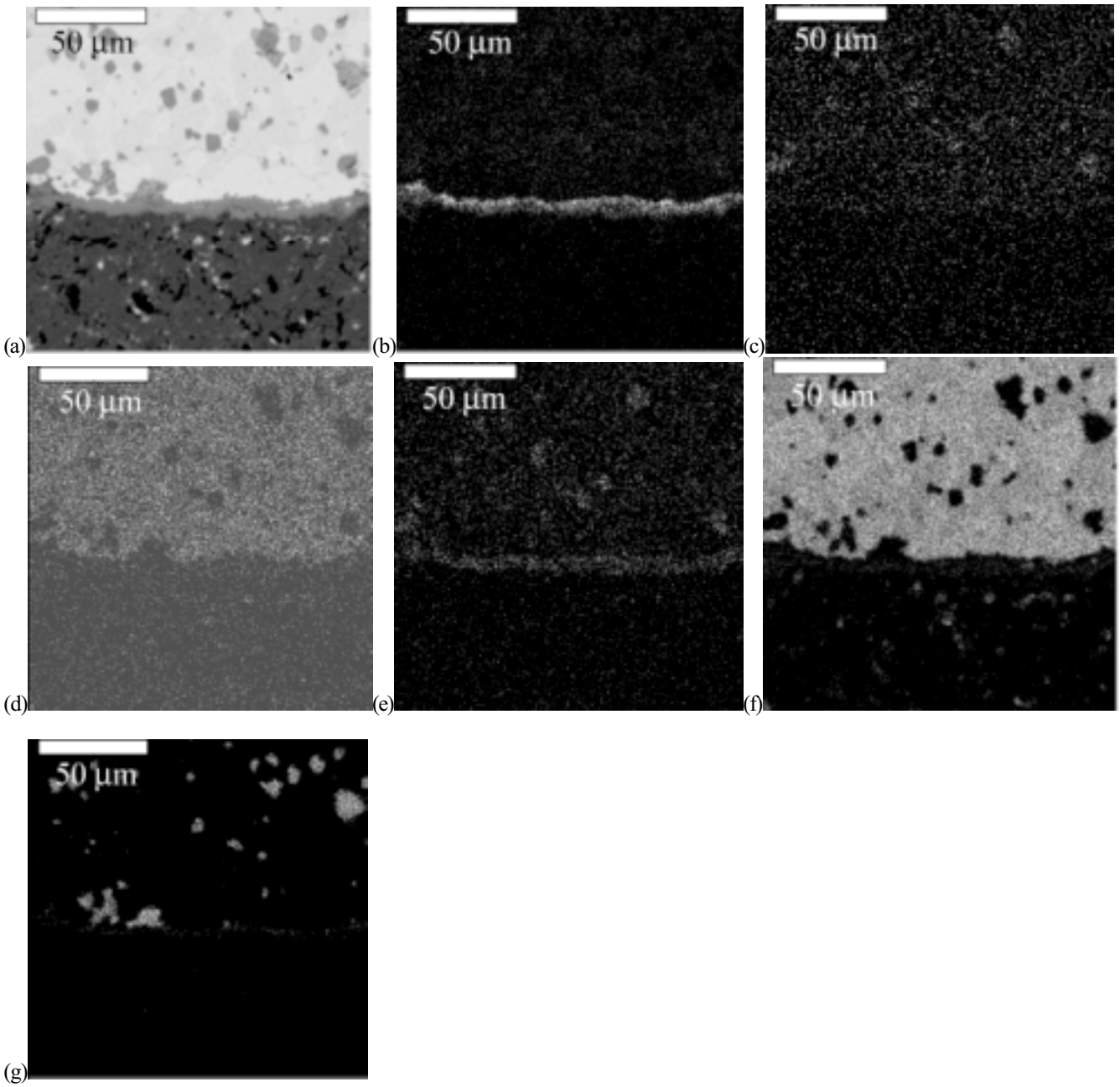
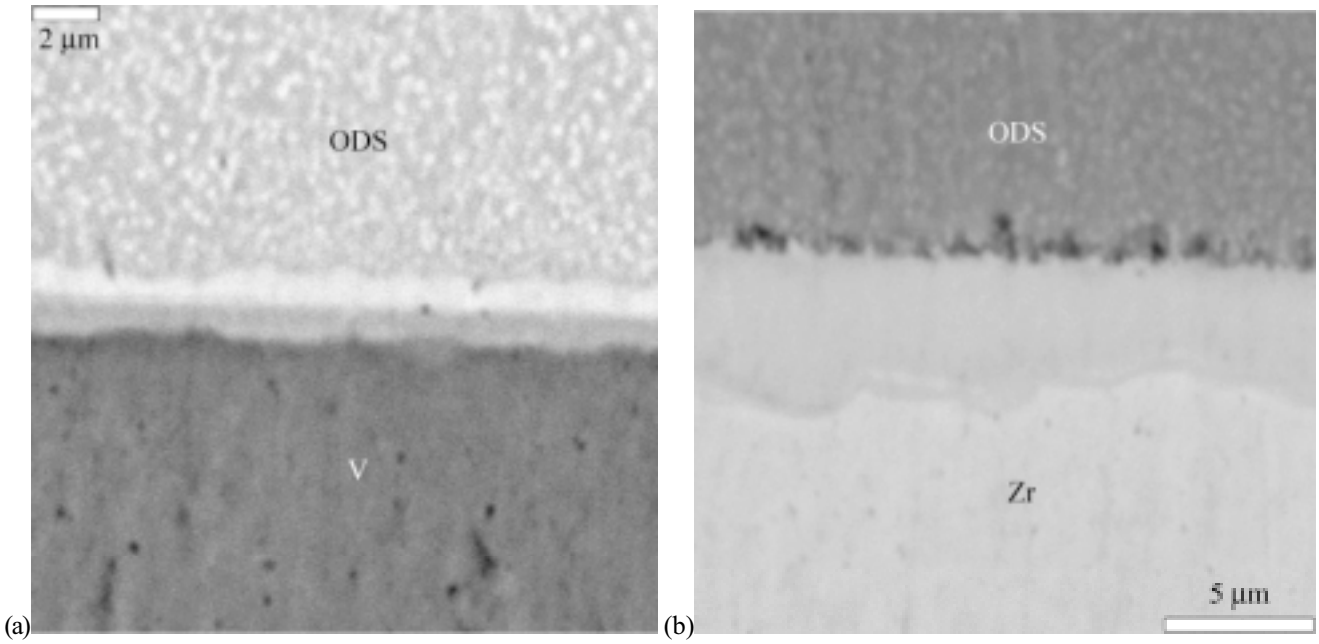
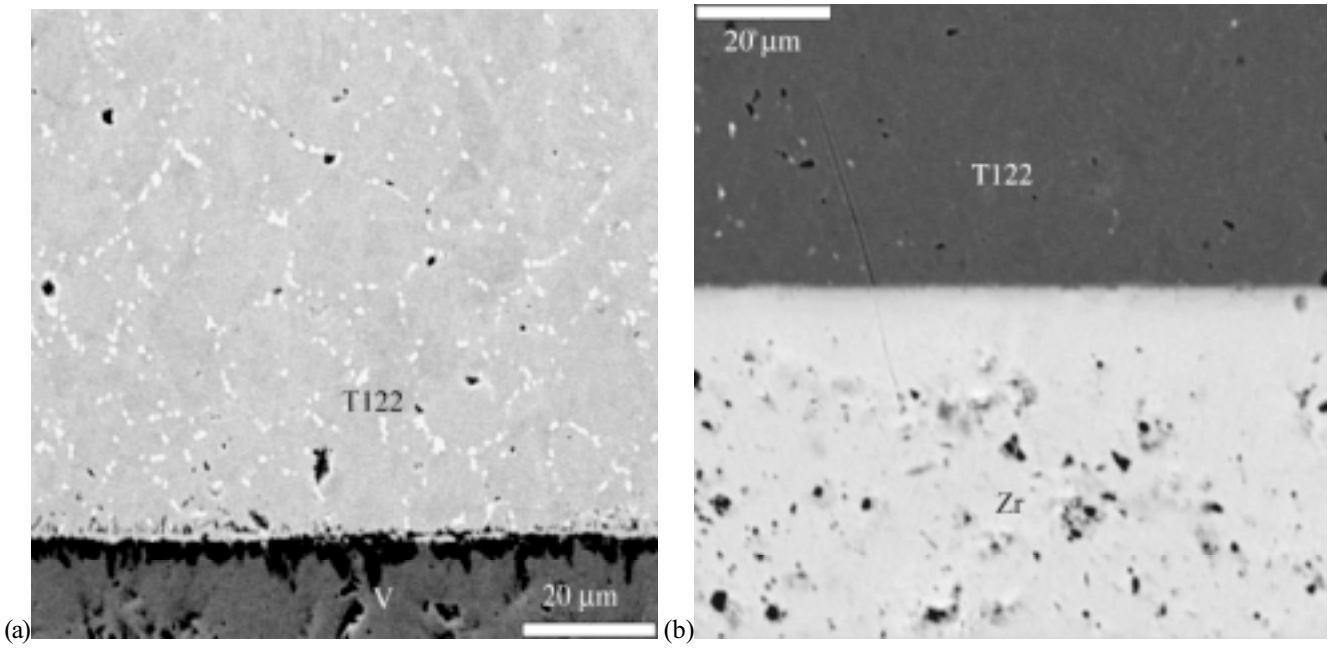


Fig. 4. Backscattered electron image (a) and X-ray maps for (b) Mo, (c) Ru, (d) U, (e) Zr, (f) Pu, and (g) Nd for the Fuel versus V couple.



(a) (b)

Fig. 5. SEM micrographs showing the diffusion structures that developed for the couples between ODS cladding alloy 1 and V and Zr after annealing at 700˚C for 50 hours.



(a) (b)

Fig. 6. SEM micrographs showing the diffusion structures that developed for the couples between the HCM12A cladding and V and Zr after annealing at 700˚C for 200 hours.

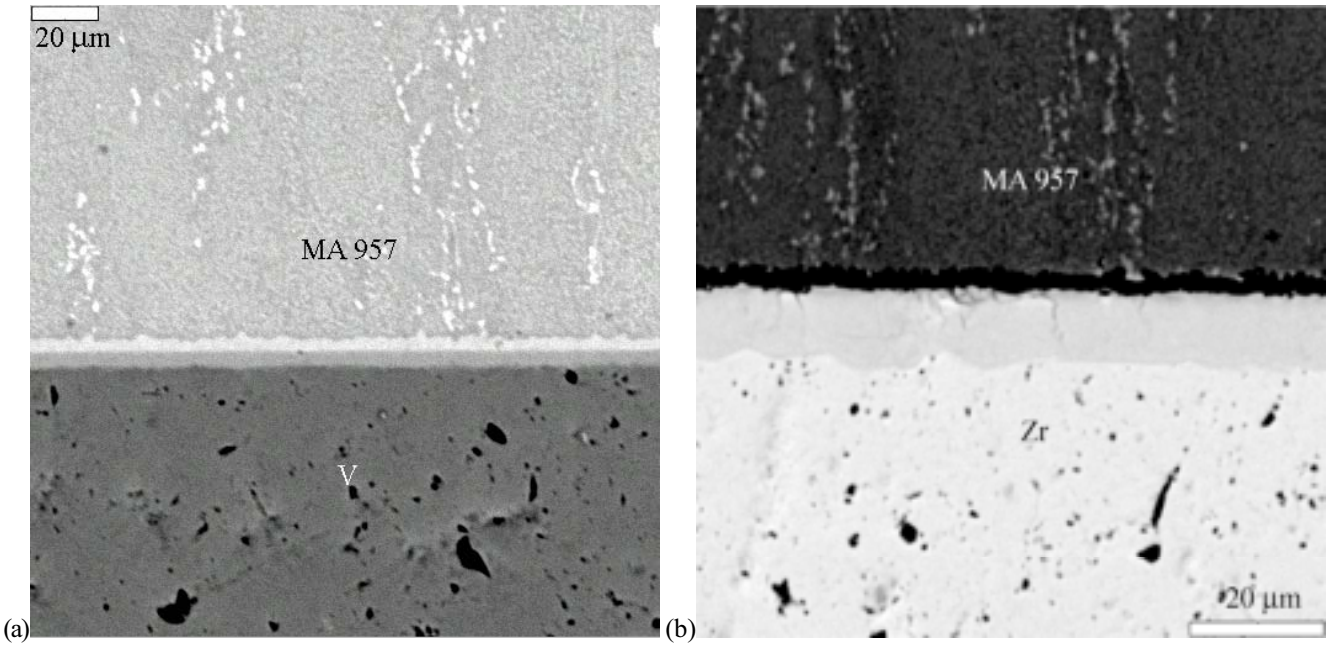


Fig. 7. SEM micrographs showing the diffusion structures that developed for the couples between the MA957 cladding and V and Zr after annealing at 700˚C for 200 hours.